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**MULTISCALE CHARACTERIZATION TECHNIQUE FOR  
CHEMICALLY REACTIVE MULTIPHASE COMPOSITE  
MICROSTRUCTURES (PREPRINT)**

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# Multiscale characterization technique for chemically reactive multiphase composite microstructures

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## ABSTRACT

The objective of this article is to outline a technique for characterizing the influence of spatial heterogeneity of two reactant phases in a three-phase chemically reactive composite material microstructure. Here, we describe a computational technique that calculates the *reaction yield* for multiple microstructure length scales based on the spatial relationship between the two reactive phases and the reaction stoichiometry. The *reaction yield* is a quantitative metric that characterizes the fraction of the chemical reaction that proceeds based on dividing the bulk microstructure into sub-regions of a particular length scale. The results of using this technique show how the reaction yield can be used to quantify differences in microstructure related to particle size and area fractions of the reactant phases. For example, we show that a bimodal microstructure has a higher reaction yield at every length scale when compared to a monomodal microstructure. The reaction yield can be an important metric for characterizing the spatial heterogeneity between two reactive phases in a three-phase composite, which can be important for tailoring the material microstructure for bulk reactivity.

KEYWORDS: Multiscale characterization; three-phase microstructure

## INTRODUCTION

When assessing microstructure-property relationships for various materials, quantitative microstructure descriptors are required to sufficiently characterize the microstructure at length scales relevant to bulk behavior. For instance, in 2-phase composite microstructures, the microstructure metrics typically relate to the size distribution, aspect ratio, alignment, and clustering of second phase particles and are calculated through a number of different quantitative stereological techniques, such as two-point correlation functions or lineal path functions. The ability to characterize materials through microstructure descriptors allows researchers to correlate the microstructure to macroscopic properties and then engineer the material microstructure through manipulating processing, thereby completing the processing-structure-property cycle.

The open literature contains a wealth of studies pertaining to the influence of the second reinforcement phase on macroscopic properties of two-phase composites. However, an important class of materials exist that contain multiple, distinct phases, the so-called *multi-phase composites*. Spatial heterogeneity in these systems is defined by a combination of the spatial heterogeneities of the different reinforcing phases. It is the purpose of the present work to quantify the combined effects of spatial heterogeneity of each reinforcement phase on the overall spatial heterogeneity of the multi-phase composite microstructure. Since multi-phase composites are often designed to be multi-functional materials, it is expected that the overall spatial heterogeneity of the composite will have complex and/or coupled effects on the resulting mechanical and physical properties. One such example of a multi-phase composite is a three-phase composite with two reinforcement phases that react chemically. The spatial arrangements of the individual particulate reinforcements will have an effect on the mechanical behavior of the composite. Moreover, the overall spatial heterogeneity of two phases will have an effect on the bulk reactivity of the material, since both phases must be present in order for the reaction to proceed. The present article seeks to quantify this spatial heterogeneity using an extension of the homogeneous length scale analysis of Spowart et al [2]. The computationally-efficient Multi-Scalar Analysis of Area Fractions (MSAAF) technique is used to compute the local area fraction statistics of both the individual reinforcement phases and the reactivity over multiple length

scales, in an attempt to provide a general framework for quantifying the effect of spatial heterogeneity on the reactivity of particle-reinforced composites.

The objective of this article is to outline a technique for characterizing the influence of spatial heterogeneity of two reactant phases in a three-phase chemically reactive composite material microstructure. This article is organized as follows. Section 2 describes the random sequential adsorption technique used for generating synthetic two-phase and three-phase composite microstructures. Section 3 describes the MSAAF technique for two-phase microstructures and its extension to three-phase chemically reactive microstructures. Section 4 shows the results of applying this technique to three-phase microstructures of varying area fractions and sizes. Section 5 presents conclusions of this work.

## 1. METHODOLOGY FOR SYNTHETIC MICROSTRUCTURE GENERATION

The three-phase composite microstructures considered in the present work comprise two different circular reinforcing phases (A and B) with area fractions  $\phi_A$  and  $\phi_B$ , which reside within a single matrix phase. Furthermore, the two reinforcing phases are simulated as either monomodal (i.e.,  $d_A = d_B$ ) or bimodal (i.e.,  $d_A \neq d_B$ ). The two-dimensional packing efficiencies of monomodal and bimodal reinforcements are different; therefore it is important to make this differentiation when comparing microstructures with equivalent area fractions.

In these simulations, a random sequential absorption (RSA) algorithm [5, 6] was used to place two different particle types within the simulation domain. The microstructure domain size is 4096 pixels x 4096 pixels for this work. Figure 1(a) shows a 1024 x 1024-pixel sub-region of a RSA-generated two-phase microstructure with area fraction of  $\phi = 0.50$  and diameter of  $d = 42$  pixels, which was generated using a MATLAB synthetic microstructure generator [8] based on Tschopp et al [1]. This MATLAB code was modified for three-phase microstructures. A random-number generator was used to decide the order of placing each phase, and particles were placed sequentially until the required area fraction of each phase was reached. The influence of boundary effects was mitigated by imposing periodic boundary conditions on the edges of the microstructure domain; particles that extended beyond the bounds were wrapped back onto the

opposite side of the domain. For the three-phase (monomodal) composite microstructures, three different area fractions were used for each phase ( $\phi_A$ ,  $\phi_B = 0.10, 0.20$ , and  $0.30$ ), creating six unique configurations. In this case, both phases have a diameter of  $d_A = d_B = 42$  pixels. Figure 1(b) shows a sub-region of a RSA-generated three-phase microstructure with  $d_A = d_B = 42$  pixels and  $\phi_A = 0.30$  (red),  $\phi_B = 0.20$  (black). Generating a synthetic microstructure with  $\phi_A = 0.30$  and  $\phi_B = 0.30$  was unsuccessful; typically RSA-generated microstructures with combined area fractions ( $\phi_A + \phi_B$ ) above  $0.54$  are unlikely to complete due to their exceeding the 2-D jamming limit for monosized disks [9].

The modified RSA algorithm was also used to create three-phase bimodal composite microstructures by placing two different-sized populations of particles within the simulation domain. As with the monomodal composite microstructures, the bimodal microstructures used three different area fractions for each phase as well:  $\phi_A$ ,  $\phi_B = 0.10, 0.20$ , and  $0.30$ . However, in these microstructures, the first phase ( $\phi_A$ ) was assigned a diameter of  $d_A = 14$  pixels, while the second phase was assigned a diameter of  $d_B = 42$  pixels. The algorithm was adjusted so that the larger particles were always placed in the simulation domain first, and the smaller particles last, in sequential fashion, until the desired area fraction of each phase was reached. Again, periodic boundary conditions were used to mitigate any possible boundary effects. Note that the  $\phi_A = 0.30$ ,  $\phi_B = 0.30$  microstructure was successful due to the more efficient packing of bimodal particles. Figure 1(c) shows a sub-region of a RSA-generated three-phase microstructure with  $d_A = 14$  pixels,  $d_B = 42$  pixels and  $\phi_A = 0.30$  (gray),  $\phi_B = 0.20$  (black). For bimodal cases, the limiting area fraction is higher than the mono-sized disk 2-D jamming limit [10], since this is the minimum bound. It is anticipated that the limiting area fraction for bimodal disk packing using the RSA algorithm is a strong function of the ratio between particle diameters. This phenomenon has a physical basis in that for certain size ratios, the smaller particles have the ability to more efficiently pack inside the interstices formed between the larger packed particles.

## 2. MULTISCALE CHARACTERIZATION TECHNIQUE

### 3.1 MSAAF technique for two-phase microstructures

In previous work [1, 2], the multiscale analysis of area fractions (MSAAF) technique has been used to quantify the homogeneous length scale ( $L_H$ ) of a real or synthetic microstructure as a function of second phase area fraction, aspect ratio, and particle alignment. The homogeneous length scale is a scalar metric associated with the phase homogeneity within a microstructure, which was developed to quantify a representative length scale for a particular microstructure based on the variations in area fraction of a particular phase as a function of length scale ( $Q$ ). Qualitatively,  $L_H$  is the length scale at which the variation between each sub-region becomes indistinguishable from a larger area of material. In other words,  $L_H$  is the minimum length scale necessary to construct a volume element representative of a particular microstructure to a given degree of statistical confidence, typically selected to be 99.0% in the isotropic case.

The multiscale analysis of area fractions (MSAAF) operates in the following manner [1]. First, a digitized microstructure is re-sampled at various resolutions (corresponding to various length scales,  $Q_n$ ) to determine the area fraction of a particular sub-region. For example, in Fig. 2, the microstructure shown in Fig. 1(a) is subdivided into 3 lower resolutions/length scales:  $Q_1$ ,  $Q_2$ , and  $Q_3$ . After subdivision, the effective area fraction ( $A_f$ ) is assigned to each sub-region, visually depicted as the gray level in Fig. 2. By using all sub-regions available at a chosen length scale, an average area fraction and standard deviation ( $\zeta_{A_f}$ ) in area fraction is determined for that  $Q_n$ . Though the average area fraction over all sub-regions for a particular length scale will not change with length scale, the standard deviation over all sub-regions will.

Consequently, the principal result of this technique is a plot depicting the evolution of the coefficient of variation ( $\psi$ ) - the ratio of standard deviation in second-phase area fraction to the global area fraction,  $\zeta_{A_f} / A_f$  - as a function of  $Q$ . Such a plot for the  $\phi = 0.50$ ,  $d = 42$  microstructure is shown in Fig. 3. When this plot decreases to a specified level of confidence ( $\psi = 0.01$ ), the corresponding  $Q$  is termed the homogenous length scale ( $L_H$ ). Differences in homogeneity between isotropic microstructures can be characterized using a value of  $L_H$  for each microstructure at a predetermined level of confidence [1, 2]. For this reason, we use the isotropic MSAAF analysis of synthetic microstructures to characterize homogeneity with the 1% homogenous length scale,  $L^{0.01}_H$ , i.e., the scale at which the variation in microstructure sub-region area fraction becomes less than 1% ( $\psi \leq 0.01$ ).

### 3.2 MSAAF technique for three-phase microstructures

This technique can be extended to composite materials that contain any number of different particle types (*multiphase* composites) with either discrete or continuous distributions of particle size (*polydispersivity*). Here we extend the work to three-phase composites with a matrix phase and two phases (reactants) that chemically react to produce a reactant product and heat, since chemical synthesis reactions are typically exothermic. In this hypothetical material, the proximity of the two reacting phases in the appropriate stoichiometric ratio will play an important role in the bulk reactive properties of the three-phase composite. Following the MSAAF technique, this characterization technique is termed the multiscale analysis of area fractions for chemical reactivity (MSAAF-CR), as it quantifies the reaction yield at each length scale based on the stoichiometry of the reactants within each sub-region.

The multiscale analysis of area fractions for chemical reactivity (MSAAF-CR) operates in the following manner. First, a digitized microstructure is separated into two images which contain the reactant phases, A and B. As in the MSAAF technique, these two images are re-sampled at various resolutions (corresponding to various length scales,  $Q_n$ ) to determine the area fraction of reactant within a particular sub-region (visually depicted as the gray level in Fig. 4). It is then necessary to determine the amount of the chemical reaction that proceeds based on the area fractions and stoichiometry of the reacting phases within each sub-region. For instance, the general form for a direct combination reaction like that discussed here is,



where  $x$  and  $y$  represent the amounts of A and B (in moles)<sup>1</sup> required to form the reactant product  $A_xB_y$ . There may be a number of possible reactions between A and B. For the purposes of this paper, here we consider  $x = 1$  and  $y = 1$ , i.e.,  $A + B \rightarrow AB$ . Now, the local regions of microstructure within each sub-region will contain varying amounts of A and B, which will

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<sup>1</sup> Since the chemical reaction units are in moles and the amounts of A and B from an image will be in units of area (volume in 3D), there will be a constant for each reactant relating the area (volume) to the number of moles. To simplify, here we have used 1 pixel of A or B is equal to 1 mole of A or B.



determine the degree to which the reaction proceeds to completion in each sub-region. In general, for sub-regions containing  $\alpha$  Ni and  $\beta$  Al where the stoichiometric ratios are not precisely met, there will either be excess A or B left over at the end of the reaction, and the fraction of reaction product will be less than that determined by the stoichiometric values at the bulk length scales. The general reaction will follow the form,



There are two possible scenarios, depending on how the stoichiometric ratio,  $R = \beta/\alpha$ , within each sub-region compares with the ideal stoichiometric ratio,  $R_0 = y/x$ . For each scenario, the amount of product is determined by the limiting reactant. For both scenarios, the amounts of reactant product,  $\gamma$ , and excess reactants,  $\delta$  or  $\varepsilon$ , are given by:

$$\gamma = \begin{cases} R \geq R_0 & \alpha/x \\ R < R_0 & \beta/y \end{cases}, \quad \delta = \begin{cases} R \geq R_0 & 0 \\ R < R_0 & \alpha - \beta/R_0 \end{cases}, \quad \varepsilon = \begin{cases} R \geq R_0 & \beta - \alpha R_0 \\ R < R_0 & 0 \end{cases} \quad (3)$$

Using Eq. (3), the amount of reaction that proceeds for each sub-region can be calculated as well as the amount of excess reactant within each sub-region. The evolution of both quantities with increasing length scale may be important for characterizing the influence of the three-phase composite material microstructure on reactive properties in the bulk material.

Figure 4 shows the influence of length scale on the hypothetical reaction given in Eq. (1) with  $R_0 = 1$ . The first and second columns of images show the evolution of area fraction for A and B as a function of length scale, as in Fig. 2. The third and fourth column of images shows the evolution of the amount of reaction product ( $\gamma$ ) and excess reactant ( $\delta$  or  $\varepsilon$ ) as a function of length scale. Notice that at smaller length scales, the general trend is for the amount of the reaction product to be small and for the excess reactants to be large, since most sub-regions do not contain both of the necessary reactants. However, as the length scale of the sub-regions increases, the amount of reaction product increases and the amount of excess reactants decreases as the sub-region contains more of A and B. As the length scale increases to infinity, the

reaction yield and excess reactants will be determined by the stoichiometry of the reactants in the bulk microstructure.

### 3. MSAAF RESULTS FOR SYNTHETIC THREE-PHASE MICROSTRUCTURES

The amount of reaction that proceeds in the three-phase reactive composite material can now be evaluated as a function of the length scale of the microstructure. For the following section, we use a stoichiometric ratio of  $R_0 = 1$ . The total amount of reactant product formed is the sum of the reactant product over all sub-regions for a particular length scale. The total reactant product can then be normalized by the total amount of reactant product at the bulk length scale to give a *reaction yield* for each length scale. The *reaction yield* is a quantitative metric that characterizes the fraction of the chemical reaction that proceeds based on dividing the bulk microstructure into sub-regions of a particular length scale.

Figure 5 shows a plot of the evolution of the reaction yield as a function of length scale for the 30%A-20%B monomodal and bimodal microstructures, which are shown in Figs. 1(b) and 1(c), respectively. Since the area fractions of A and B are equivalent in both distribution, this figure shows that the reaction proceeds further in the bimodal distribution than the monomodal distribution at all length scale. By using smaller particles for A, the probability of finding A and B within each sub-region was increased, enabling more of the reaction to occur at every length scale. Also, the intersection of the curve<sup>2</sup> with reaction yields of 0.05, 0.50, and 0.95 can be used to quantitatively characterize the reaction length scales (denoted as  $RY_{0.05}$ ,  $RY_{0.50}$ , and  $RY_{0.95}$ , respectively). The length scale  $RY_{0.05}$  may be an important metric for characterizing the sensitivity of the reaction to the microstructure, while  $RY_{0.95}$  may be an important metric for characterizing the complete reaction of the reactants. These metrics should be a function of the aspect ratio, area fractions, spatial relationships (i.e., clustering), and size distributions of the reactants. The ability to quantify the microstructure in such a manner may allow researchers to tailor the processing of the material microstructure for improved reactivity.

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<sup>2</sup> A cubic spline fit was used to interpolate between data points.

As an additional example, Figure 6 shows the difference in the reaction yield as a function of length scale for varying area fractions of B ( $\phi_B = 0.10, 0.20, 0.30$ ) and sizes of B ( $d_B = 7, 21$ ) for a microstructure with  $\phi_A = 0.10$ . As expected, increasing the area fraction of B allows the reaction to proceed further at every length scale, i.e., the probability of finding B within sub-regions containing A was increased with increased loading of B within the microstructure. Additionally, as in Fig. 5, the bimodal microstructure has higher reaction yields than monomodal microstructures of the same area fraction for every length scale. Interestingly, the amount of reaction that occurs at the 16-pixel length scale in the bimodal material with  $\phi_A = 0.10$  and  $\phi_B = 0.10$  is equivalent to that in the monomodal material with  $\phi_A = 0.10$  and  $\phi_B = 0.30$  ( $RY_{0.05}$  is very similar). However, at larger length scales, the monomodal material clearly reacts more than the bimodal material in this example, as the larger area fraction of B eventually contributes more towards full reaction with A ( $RY_{0.95}$  is very different). Different length scales associated with the reaction yield curve may be more important depending on the application and material.

#### 4. CONCLUSIONS

We have demonstrated a technique that can be used to identify microstructural parameters related to the chemical reactivity of heterogeneous three-phase composite materials. While this technique has been used on idealized synthetic microstructures (Fig. 1) with components A and B (reactants) which react to form compound AB in a reaction that is typically exothermic, it is applicable to real chemical reactions and experimentally-obtained microstructures. The reaction yield is a microstructure-based metric that accounts for both the amount of reactants and the reaction stoichiometry to quantify the degree of the reaction that proceeds within each microstructural sub-region. In this article, the MSAAF-CR technique has been used to show the influence of area fraction and particle size (monomodal vs. bimodal sizes) on the reactivity as a function of length scale (Figs. 5 and 6).

The ability to characterize the microstructural variability in three-phase composite materials is the first step towards correlating the relationship between microstructure and chemical reactivity in these systems. A better understanding of the microstructure-property relationship can enable

tailoring the three-phase composite microstructure for properties and performance through material processing.

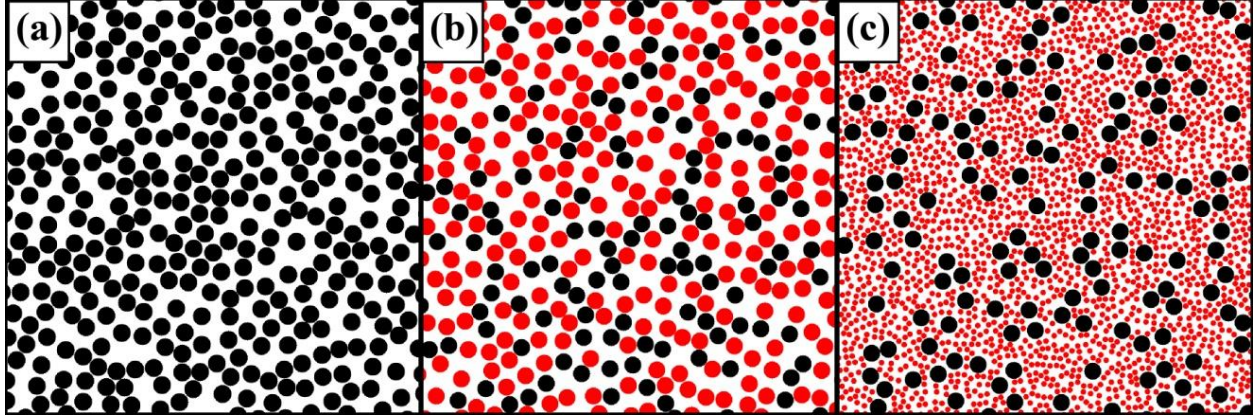
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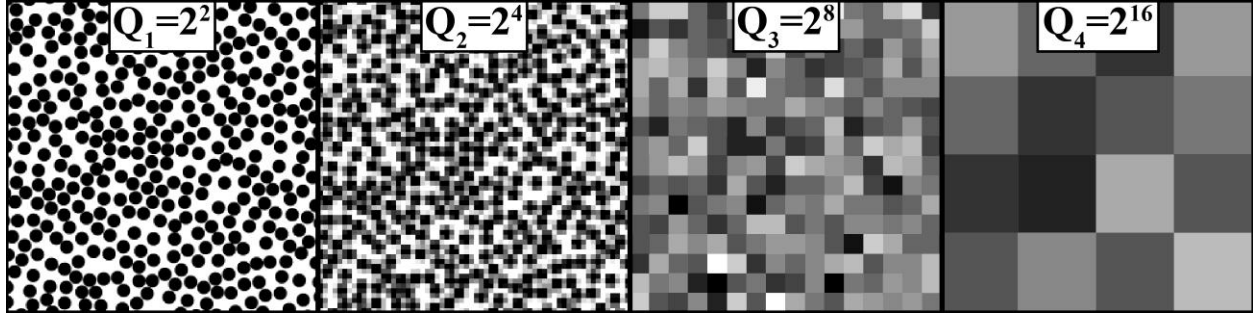
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## Figure Captions

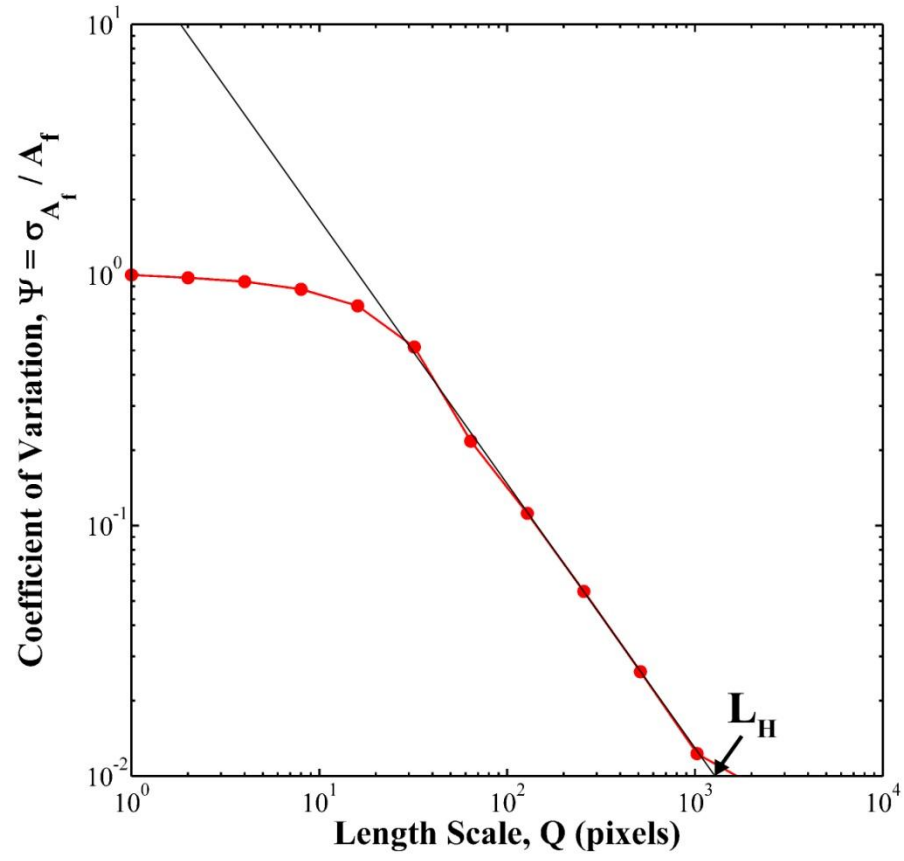
**Figure 1.** Sub-regions (1024 pixel x 1024 pixel) of various microstructures generated using the random sequential adsorption technique: (a) a two-phase microstructure with  $\phi=0.50$ ,  $d = 42$  pixels, (b) a three-phase monomodal ( $d_A = d_B = 42$  pixels) microstructure with  $\phi_A = 0.30$  and  $\phi_B = 0.20$ , and (c) a three-phase bimodal ( $d_A = 14$  pixels and  $d_B = 42$  pixels) microstructure with  $\phi_A = 0.30$  and  $\phi_B = 0.20$



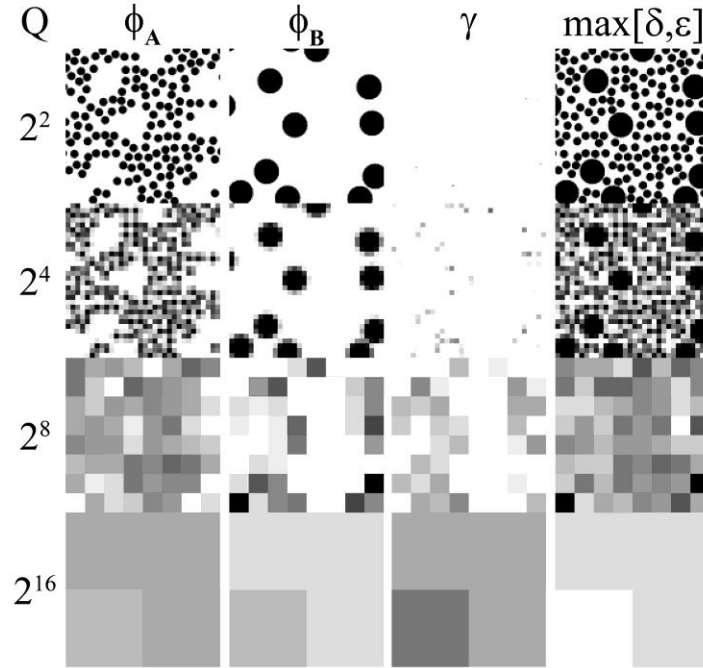
**Figure 2.** A series of images showing the evolution of the area fraction of second phase (depicted by gray level intensity) as a function of length scale,  $Q$ , according to the isotropic MSAAF technique [1, 2]



**Figure 3.** Isotropic MSAAF plot showing the evolution of the coefficient of variation as a function of length scale for the  $\phi=0.50$ ,  $d = 42$  pixel two-phase microstructure (Fig. 1(a))

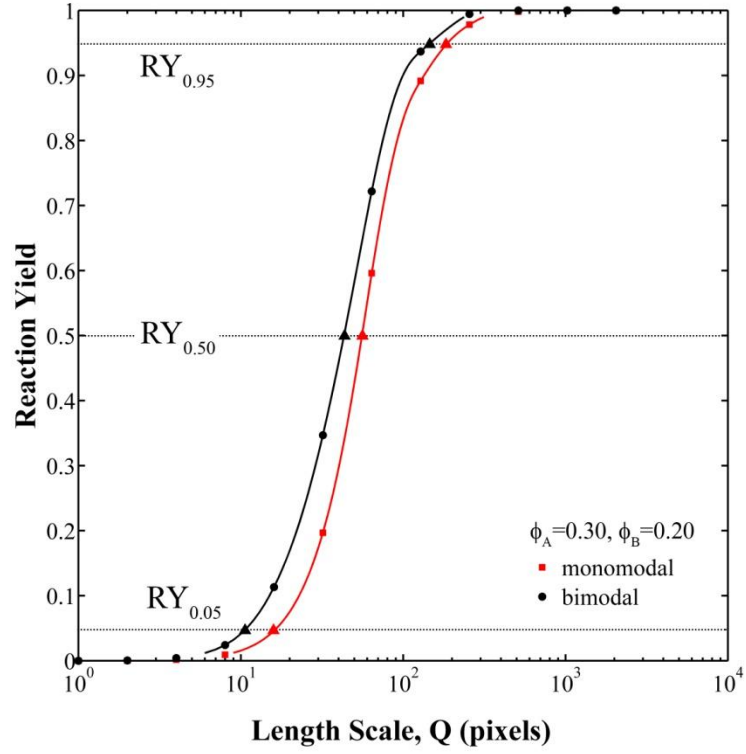


**Figure 4.** A series of images showing the evolution of the area fractions of A and B, the reaction product ( $\gamma$ ), and unreacted fraction ( $\delta, \epsilon$ ) as a function of length scale,  $Q$ , according to the three-phase MSAAF technique





**Figure 5.** Plot showing the evolution of the reaction yield as a function of the length scale for a monomodal ( $d_A = d_B = 42$  pixels) and bimodal ( $d_A = 14$  pixels and  $d_B = 42$  pixels) three-phase composite with  $\phi_A = 0.30$  and  $\phi_B = 0.20$



**Figure 6.** Plot showing the evolution of the reaction yield as a function of the length scale for monomodal ( $d_A = d_B = 42$  pixels) and bimodal ( $d_A = 14$  pixels and  $d_B = 42$  pixels) three-phase composites with  $\phi_A = 0.10$  and  $\phi_B = 0.10, 0.20$ , and  $0.30$

